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Remarks

Claims 84-93 are pending in the Application.

Claims 84-88, and 90-93 stand rejected.

Claim 89 is objected to.

Claim 85 is cancelled herein.

I. REJECTION UNDER 35 U.S.C. § 112, ¶ 2

Examiner has rejected Claim 88 under 35 U.S.C. § 112, ¶ 2, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. Examiner contends that is indefinite as to what is included under the limitation “cable-like.” Paper No. 10, at 2. Applicant traverses this rejection.

Applicant respectfully points out that “cable-like” refers to a particular composite arrangement comprising continuous carbon fibers of single-wall carbon nanotubes and is described in the Application, page 46, lines 15-17, and illustrated in Fig. 12.

As a result of the foregoing, Applicant respectfully requests that the Examiner withdraw the rejection of Claim 88 under 35 U.S.C. § 112, ¶ 2, as being indefinite.

II. REJECTIONS UNDER 35 U.S.C. § 102

Examiner has rejected Claims 84-88 and 90-93 under 35 U.S.C. § 102(a) as being anticipated by Kiang *et al.*, “Carbon Nanotubes with Single-Layer Walls,” Carbon, 33(7), pp. 903-914, 1995 (“Kiang”). Paper No. 10, at 3. When doing so, the Examiner has relied upon Zhang *et al.*, “Microscopic structure of as-grown single-wall carbon nanotubes by laser ablation,” Philosophical Magazine Letters, 78(2), pp. 139-144, 1998 (“Zhang”) purportedly to show a “state of fact.”¹ *Id.*

¹ There are only three instances under which a second reference can be used when making a §102 rejection. See M.P.E.P. § 2131.01. The only one possibly pertinent here is the third instance, namely to “[s]how that a characteristic not disclosed in the reference is inherent.” *Id.*

As to Claims 84-85, Claim 85 has been cancelled, and Claim 84 has been amended to include the limitations of cancelled Claim 85. Namely, Applicant has added the limitation to Claim 84 that the continuous carbon fiber comprises a substantial portion of single-wall carbon nanotubes having a homogeneous characteristic selected from the group consisting of lengths, diameters, helicities and combinations thereof.

Regarding Claims 84-85, Examiner contends that, because *Kiang* teaches that single-walled nanotubes tend to aggregate into bundles, the nanotubes within said bundle running substantially parallel to one another, and because “Zhang teaches that the tubes have a homogeneous diameter and are packed into a two-dimensional triangular lattice,” it is inherent to the bundled single-walled nanotubes [of *Kiang*] that they have a homogeneous diameter. Paper No. 10, at 3.

Regarding Claims 84-85 and § 102, where anticipation requires each and every claim to be found within the cited prior art reference, *Kiang* does not include the required limitation of any homogeneous characteristic stated above. Furthermore, regarding the argument that single-wall carbon nanotubes have inherently homogeneous diameters when aggregated in bundles is incorrect. The diameter homogeneity of the single-wall carbon nanotubes is highly dependent on the synthesis process. The nanotubes of *Kiang* and *Zhang*,² (a non-prior art reference) are made by processes that, not only differ substantially from each other, but also differ from the processes employed to make the continuous carbon fibers of the present invention. Additionally, reports exist showing ropes of single-wall carbon nanotubes in which the nanotubes within the rope have diameters that vary widely. See Nikolaev *et al.*, “Diameter doubling of single-wall nanotubes,” Chemical Physics Letters, 266 (5-6), pp. 422-426, 1997, Fig. 2 (Exhibit A). Thus, the assertion of inherent homogeneity of diameter of the *Kiang* nanotubes is without basis.

² *Zhang* was published in the Philosophical Magazine Letters in 1998. The present Application is a division of co-pending prior application Serial No. 10/000,746, filed on November 30, 2001, which is a continuation of prior application Serial No. 09/242,040 filed on September 13, 1999, which is the 35 U.S.C. § 371 national application of International Application Number PCT/US97/13896 filed on August 8, 1997, which designated the United States, claiming priority to provisional U.S. patent application Serial Number 60/023,732 filed on August 8, 1996. Thus, putting aside any benefits this Application receives due to its provisional application, this Application has at least an effective filing date of August 8, 1997. Accordingly, *Zhang* is not prior art.

Regarding Claim 86, Examiner contends that “it is not explicitly taught that the individual single-wall nanotubes in a bundle have homogenous lengths or helicities in any given region of the bundle. However, it is expected that at least two adjacent tubes will have the same helicity or the same length due to corresponding growth conditions.” Paper No. 10, at 3.

The speculation or expectation that single-wall carbon nanotubes within a rope would have homogeneous lengths and/or helicities is also without support. *Kiang* does not address lengths or helicities; and, Applicant notes, *Zhang* does not either. It cannot follow that such features are necessarily present; and thus such features are not inherent in *Kiang*. See *Continental* 948 F.2d at 1269, 20 U.S.P.Q.2d at 1749.

Regarding Claims 87-88, Examiner contends that “no difference is seen between the bundles of single-walled nanotubes of *Kiang et al.* and the ‘cable-like’ fibers formed from carbon fibers, each of which comprises single-walled nanotubes in a parallel orientation.” Paper No. 10, at 3.

Applicant respectfully points out that cable-like fibers are described in the present Application (page 46, lines 15-20, and Fig. 12) as being a type of composite comprising continuous carbon fibers, which themselves comprise single-wall carbon nanotubes. *Kiang* (and *Zhang*) merely teaches ropes (bundles) of single-wall carbon nanotubes—not composite structures.

Regarding Claim 90, Examiner contends that “it is not explicitly taught that the bundles of single-walled nanotubes may contain a portion that is not parallel. However, it is expected that this be the case because *Zhang* holds that a large bundle can split into sub-bundles, and additionally because *Kiang* observes this phenomenon (Fig. 2c).” Paper No. 10, at 4.

As with Claims 87-88 above, Claim 90 is directed to a composite. *Kiang* (and *Zhang*) neither teach nor suggest such a composite. That *Kiang* (and *Zhang*) teach large bundles (ropes) splitting into different directions as “sub-bundles” is not relevant because they are not composite structures as described in the present invention.

Regarding Claims 91-93, Examiner has not indicated specifically how these are anticipated by *Kiang*. Applicant respectfully points out, however, that these Claims are directed

toward molecular arrays, a molecular array being “a substantially two-dimensional array made up of single-wall carbon nanotubes aggregating (*e.g.*, by van der Waals forces) in substantially parallel orientation to form a monolayer extending in directions substantially perpendicular to the orientation of the individual nanotubes.” Application, page 38, lines 27-30. Furthermore, the molecular arrays in each of Claims 91-93 require single-wall carbon nanotubes to be aggregated in an orientation for growing a continuous carbon fiber of the present invention. *Kiang* does not teach a molecular array as described above, nor does it teach an orientation for growing any type of carbon fiber.

As a result of the foregoing, Applicant respectfully requests that the Examiner withdraw the rejection of Claims 84, 86-88, and 90-93 under 35 U.S.C. § 102(a) as being anticipated by *Kiang*.

III. REJECTIONS UNDER 35 U.S.C. § 103

Examiner has rejected Claims 86 and 90 under 35 U.S.C. § 103(a) as being obvious over *Kiang* with *Zhang*. Paper No. 10, at 3.

As an preliminary point pertaining to this § 103(a) rejection, it is unclear to Applicant whether the Examiner is combining the two references when making this rejection or the Examiner is relying only *Kiang* for this obviousness rejection and is utilizing *Zhang* merely to show that *Kiang* inherently has certain features.

To the extent the Examiner is asserting the former, Applicant respectfully points out that *Zhang* is not a prior art reference, as it was published after the effective filing date of the Application. See footnote 2 above. Thus, there is no basis for this § 103(a) rejection.

To the extent the Examiner is asserting the latter, Examiner is reminded that:

To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art and not based on applicant's disclosure.

See M.P.E.P. 706.02(j); see also *In re Vaeck*, 947 F.2d 488, 20 U.S.P.Q.2d 1438 (Fed. Cir. 1991).

With regard to Claim 86, Applicant points out that *Kiang* teaches or suggests none of the homogeneous characteristics of length, diameter, or helicity, as limitations put forth in Claim 86. As noted above, there is no teaching or suggestion (inherent or otherwise) in *Kiang* to overcome this void.

With regard to Claim 90, Applicant respectfully points out that *Kiang* teaches only ropes of single-wall carbon nanotubes—not the composite fiber comprising a plurality of continuous carbon fibers, themselves comprising single-wall carbon nanotubes.

As such there is no teaching or suggestion in *Kiang* to make the claimed combinations of Claims 86 and 90.

As a result of the foregoing, Applicant respectfully requests that the Examiner withdraw the rejection of Claims 86 and 90 under 35 U.S.C. § 103(a) as being obvious over *Kiang* with (or without) *Zhang*.

IV. OBJECTIONS

Examiner has objected to Claim 89 as being dependent upon a rejected base claim (Claim 87). Pursuant to arguments pertaining to Claim 87 above reflecting that Claim 87 is in allowable form, Applicant respectfully requests that the Examiner withdraw the objection to Claim 89.

V. **CONCLUSION**

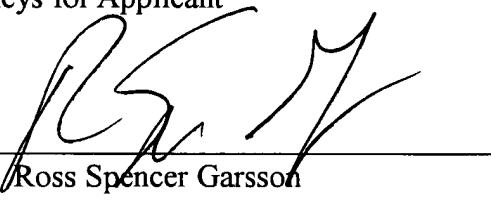
As a result of the foregoing, it is asserted by Applicant that the Claims in the Application are now in a condition for allowance, and respectfully request an early allowance of such Claims.

Applicant respectfully requests that the Examiner call Applicant's attorney at the below listed number if the Examiner believes that such a discussion would be helpful in resolving any remaining problems.

Respectfully submitted,

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Attorneys for Applicant

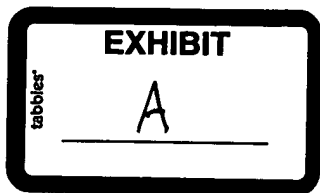
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Diameter doubling of single-wall nanotubes

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Abstract

High-yield single wall nanotube (SWNT) material consisting primarily of zero-helicity armchair tubes was annealed at 1400 or 1500°C under medium-high vacuum, flowing argon and flowing hydrogen atmospheres. As many as 60% of the nanotubes so treated coalesced with neighbors, the proportion depending on the atmosphere, resulting in nanotubes with twice (2.7 nm) and occasionally three times (4.1 nm) the diameter of the (10,10) tube that is the dominant armchair tube in these samples. Since only nanotubes with equal helical pitch angle and chirality can coalesce seamlessly, the high conversion ratio adds support to other recent evidence that no type of SWNT is formed in these samples in greater numbers than the (10,10) tube.

1. Introduction

Since their discovery in 1991 [1], carbon nanotubes have been the focus of increasing attention, spurred by their predicted, and now in some cases measured, novel materials properties. In 1993 [2], it was discovered how to produce samples containing the most basic type of nanotubes: those with only a single layer, as compared with earlier samples containing nanotubes comprising multiple concentric cylindrical layers. Single-wall nanotubes (SWNT) have several advantages over their multiwall brethren, the most notable being their lack of defects. No defect has ever been observed in any as-grown SWNT, whereas multiwall nanotubes (MWNT) invariably contain significant defects at least every few microns of length, regardless of their method of

formation. This, together with the purity of structure of SWNTs makes it almost certain that the latter will exhibit more ideal materials properties such as electrical and thermal conductivities, and strength, compared with MWNT. Another important advantage of SWNT is that their relative simplicity makes them amenable to theoretical and computational scrutiny.

Very recently, our group continued this trend of refining nanotube material [3], reporting a method, based on laser-vaporization of a graphite/metal composite target, of making high-yield samples of SWNT with an amazing uniformity of diameters (~ 1.36 nm) [3,4]. These samples consist of 'ropes' of 100–500 individual nanotubes aligned in a two-dimensional crystal packing arrangement over essentially their entire length. Electron diffraction experiments have shown that the ropes are dominated by achiral (zero helicity) armchair tubes (in which there are C–C bonds normal to the tube axis), with the rest

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consisting mainly in tubes of small helical pitch angle about the armchair configuration [5]. As discussed previously by us [3], armchair tubes are favored during growth because the carbon atoms at the open, growing edge can pair up to form only slightly strained triple bonds, thereby vastly reducing their edge energy.

Nanotube layers are denoted uniquely by the indices (m , n) to indicate how a tube is constructed from a planar graphene sheet. Armchair tubes have $m = n$, with n (triple-bonded) pairs of carbon atoms around any circumference. They are the only truly metallic SWNTs, the others being either large- or small-gap semiconductors [6]. In Ref. [3] we reported the astonishingly narrow diameter distribution observed in high-yield SWNT samples, with a quantitative fit to x-ray diffraction data yielding an average diameter of 1.36 nm, the predicted diameter of the (10,10) tube. This tube is special among armchair tubes by virtue of its high symmetry; the cap of a (10,10) tube – half of a C_{240} fullerene – has the same symmetry as C_{60} . In fact, the (10,10) tube is precisely the right size to accommodate a C_{60} molecule inside it with the appropriate van der Waals spacing.

The reason, though, that we sometimes refer to the (10,10) tube as the ‘buckyball of nanotubes’ has more to do with its dominance in SWNT samples prepared by laser vaporization than with its symmetry (although symmetry likely has a role to play in favoring the (10,10) tube over other armchair tubes). Just as C_{60} is the favored form of carbon under conditions constraining carbon cluster to remain nanoscopic, the (10,10) tube is the best solution if the carbon cluster is constrained to remain open beyond the point when it would have normally closed into a fullerene. For details of our proposed growth mechanism of (10,10) tubes, see Ref. [3].

We have built up support from several directions that the (10,10) tube is favored in our SWNT samples, including x-ray diffraction [5], electron diffraction [6], resistivity measurements [3,7] and IR/Raman spectroscopy [8], and theory [9]. In this Letter we add to this list from a new angle, reporting conditions under which individual tubes coalesce to form diameter-doubled tubes with high conversion efficiency.

Coalescence of fullerenes is not a new phe-

nomenon. The possibility was first suggested [10] in 1991 to explain the appearance of increasing amounts of $M_x@C_n$, with 2–4 metal atoms and $n > 100$, as nascent metallofullerenes react with one another between successive laser vaporization shots of a composite graphite/metal target. (As a notable aside, it was in these publications that the first speculation was made on the possibility of forming ‘bucky tubes’, prior to their observation in TEM micrographs.) Subsequently, coalescence of empty fullerenes was also reported [11]. The extension of such reactions to tubes would be inconceivable were it not for the remarkable tendency for individual tubes to align over their entire lengths [3]. However, as discussed further below, in order for tubes to coalesce, they must also have the same chirality and helical pitch angle. Since only tubes with the same pitch angle and chirality can coalesce seamlessly over great lengths, as is observed, the most likely candidates for coalescing tubes are achiral ones, most of which are already known from the evidence cited above to be (10,10) tubes.

2. Experimental

The production of high purity specimens (70–90% yield) of single-wall nanotubes by laser vaporization of metal-doped graphite rods has been described elsewhere [3,4]. Samples weighing 3–4 mg were placed in a ceramic boat in a 2-in. alumina flow tube and treated under various atmospheres at temperatures up to 1500°C within a tube furnace (Lindberg Blue M). The conditions for each experiment are summarized in Table 1.

In experiment 1, the flow tube was pumped down with a turbopump (Balzers), and the pressure measured by a hot cathode ionization gauge (Varian). A vacuum of 2 μ Torr was achieved by extensively outgassing the sample and system at 1100°C for 8 h while the rest of the vacuum system was heated with heating tape to 110°C. No tube coalescence was observed under these conditions. The furnace temperature was subsequently raised to the final 1400°C treatment temperature.

In experiments 2 and 3 the flow tube was filled, after outgassing, with UHP hydrogen at 6 mTorr, flowing at 2 sccm at 1400°C for 2 h, and at 1500°C

Table 1

	<i>T</i> (°C)	Flow rate (sccm)	Atmosphere	Time (h)	Coalescence (%)
1	1400	—	2 μ Torr vacuum	2	5
2	1400	2	6 mTorr H ₂	2	50
3	1500	2	6 mTorr H ₂	3	60
4	1500	300	800 Torr Ar (8 mTorr impurities)	2	45
5	1500	300	800 Torr filtered Ar (0.8 mTorr impurities)	2	30

for 3 h, respectively. Hydrogen was metered in via a high precision mass-flow controller (MKS).

In experiments 4 and 5 the flow tube was filled, after outgassing, with UHP argon at 800 Torr (5–10 ppm, or 4–8 mTorr impurities), flowing at 300 sccm. The argon flow was controlled via a float tube flow meter (Matheson). In experiment 5, the concentration of trace impurities in the argon was reduced to below 1 ppm (i.e., less than 0.8 mTorr) by an Oxyzorb catalytic filter (MG Industries).

Specimens were prepared for TEM imaging by laying a small piece of SWNT material on a lacey carbon TEM grid and then placing a drop of methanol over it. This technique yields many more cross-sectional views of ropes as they curve through the focal plane of the microscope than samples prepared by depositing on a TEM grid a drop of material sonicated in methanol, which tends to produce ropes lying flat on the grid.

3. Results

Diameter histograms from each experiment, as well as from untreated samples are shown in Fig. 1. Diameters were measured from TEM micrographs, with an accuracy of about ± 1.5 Å. In each treated case (1b–f), a bimodal distribution is evident, with one peak centered on the (10,10) tube diameter of 1.36 nm, and the other centered at about twice this diameter. The fraction of tubes coalescing in the sample held under medium-high vacuum (Fig. 1b) is very small, only about 5%. Table 1 lists the fraction of tubes coalescing for each experiment. A typical TEM micrograph of SWNT annealed in experiment 3 is shown in Fig. 2. In addition to many 1.36 nm diameter tubes, there are several that are 2.7 nm in diameter. The wider nanotubes are slightly flattened as they curve through the vertical plane, so average

diameters were determined from measurements of their circumference. A few tubes with three times the circumference of the (10,10) tube have also been observed, but they are relatively rare.

The poverty of diameter-doubling in experiment 1 (heating to 1400°C under medium-high vacuum), compared with the abundance of doubling in experiment 2 indicates that tube coalescence is activated by hydrogen. Supportive of this, the higher temperature

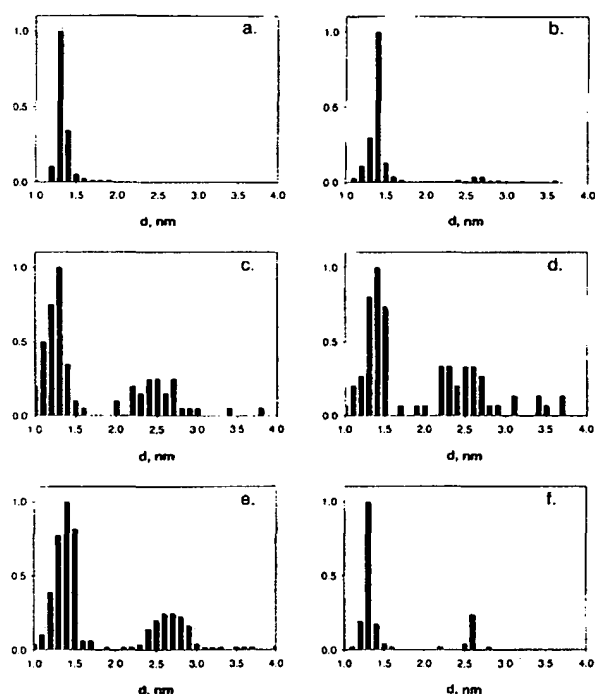


Fig. 1. Histograms of diameters for nanotubes (a) untreated (no coalescence), and (b–f) treated under various conditions: (b) baked at 1400°C under 2 μ Torr vacuum for 2 h (5% coalescence); (c) baked at 1400°C under 6 mTorr hydrogen for 2 h (50% coalescence); (d) baked at 1500°C under 6 mTorr hydrogen for 3 h (60% coalescence); (e) baked at 1500°C under 800 Torr of UHP argon (~ 8 mTorr impurities) for 2 h (45% coalescence); (f) baked at 1500°C under 800 Torr of Oxyzorb-filtered UHP argon (~ 0.8 mTorr impurities) for 2 h (30% coalescence).

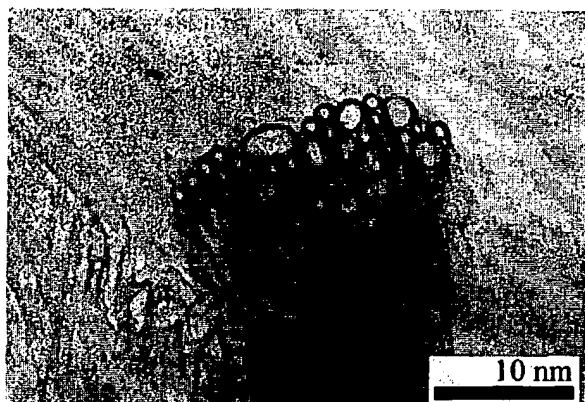


Fig. 2. Cross-sectional view of a rope as it curves through the TEM focal plane. Double-diameter tubes are slightly flattened as they bend in the vertical plane. One triple-diameter tube is also visible.

and longer exposure to hydrogen of experiment 3 compared with experiment 2, produced more coalescence. At these temperatures, equilibrium favors atomic over molecular hydrogen, so almost certainly the former is the active species. The small amount of doubling in experiment 1 is consistent with the small background pressure (2 μ Torr), most of which derives from water vapor and its various equilibrium products, including hydroxyl radicals, and atomic oxygen and hydrogen. The total vapor pressure of impurities (principally H_2O and the species, including atomic hydrogen, in equilibrium with it at the prevailing temperature) in experiment 4 are estimated to be about 8 mTorr, similar to the H_2 pressure in experiments 2 and 3, and results in a similar fraction of doubled tubes. When most of these impurities are removed with an Oxyzorb filter to a partial of 0.8 mTorr, the fraction of doubled tubes decreases accordingly.

4. Discussion

Based on these results, we propose a mechanism for SWNT coalescence in which a gas-phase species (e.g., H-atom or OH radical) attacks the side of a nanotube, breaking a C–C bond thus producing a defective site in an otherwise perfect graphene network. Either the original attacking species or the newly formed reactive carbon edge atoms can subse-

quently attack a neighboring tube, producing a defect in the second tube. Once these adjacent defects have formed, there will be a strong thermodynamic driving force, resulting from the reduced strain energy of larger diameter tubes, for the two neighboring tubes to join together if they can. While there is no kinetically facile or energetically advantageous way for two parallel tubes to connect *locally* (e.g., by forming a seamless bridge between them), propagation of the two opposing defects along their mutual lengths while zipping closed the graphene sheets in their wake should be favorable kinetically and energetically (see below). After zipping begins, the same driving force would impel the two tubes to knit together for as long as they run together. Once two tubes have begun to knit together, this process must eventually terminate when one of the tubes ends. If the second tube extends further than the first, one would anticipate an abrupt narrowing from 2.7 nm to 1.36 nm diameter; one such instance is shown in Fig. 3.

In accordance with our observation of very little coalescence of larger-diameter tubes, smaller diameter nanotubes should be more susceptible to coalescence for two reasons: (1) The reactivity of a curved graphene sheet increases as the tube diameter becomes smaller, since curvature introduces more s-orbital character into the π -orbitals. (2) The coalescence exothermicity of smaller diameter tubes is greater than for larger diameters. The strain energy of a nanotube, disregarding end caps, is $E_s = \epsilon_s L/D$ where L is the length of the tube of diameter D and

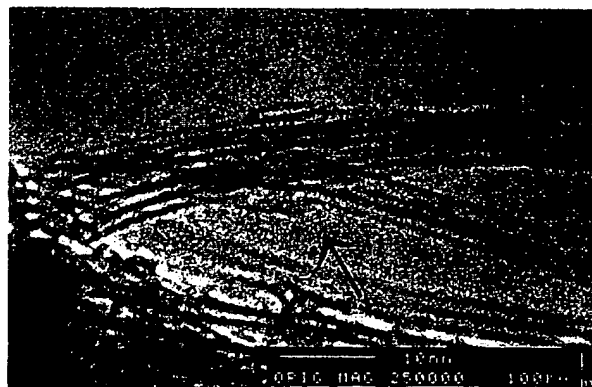


Fig. 3. Side view of a necked-down tube. The diameter narrows abruptly from 2.7 to 1.36 nm.

$\epsilon_s \approx 10$ eV [3]. For coalescence of two (10,10) tubes, the enthalpy change is therefore approximately 11 eV/nm of tube length. The enthalpy change is 30% smaller for coalescence of the resulting (20,20) tube and another (10,10) tube to yield a diameter-tripled tube, continuing to smaller exothermicities for higher-order coalescence.

For two tubes to knit together seamlessly, they must have the same helical pitch angle and chirality, otherwise they immediately lose their registry, making the distance between open edges too great to bridge. For a sample of nanotubes with random helicities it is very unlikely that these conditions will be fulfilled for many pairs of neighboring tubes. This does not preclude other types of chemistry between unmatched carbon networks, like formation of bridges between tubes, but significant numbers of coalesced tubes cannot be obtained in such a sample.

The observed high percentage of coalescing SWNT therefore argues strongly for the converse: that the starting material is dominated by nanotubes of a single helicity. The observed $\sim 60\%$ maximum conversion provides rather dramatic new evidence that at least this fraction of nanotubes have the same pitch angle and chirality. The simplest way to have both properties be the same is to be achiral, i.e., zero-helicity. These results are consistent with and supportive of other data from x-ray diffraction [3], electron diffraction [5], resistivity measurements [3,7], and IR/Raman spectroscopy [8], indicating that the high-yield SWNT samples made by laser-vaporization are dominated by the remarkable (10,10) tube.

5. Conclusion

We have found that at 1400 and 1500°C up to 60% of single-walled nanotubes coalesce, forming diameter-doubled tubes. The fraction of coalesced SWNT depends on the annealing atmosphere and is proportional to the pressure of reactive gases, most

notably atomic hydrogen. A simple model is proposed, in which coalescence is initiated when a gas-phase atom attacks two adjacent nanotube walls. Coalescence is thermodynamically driven by the reduced strain energy of the resulting larger diameter tube. Since the nanotubes must have the same helical pitch angle and chirality for coalescence to occur over significant lengths, the high fractions of doubling supports the claim that high-yield laser-vaporization SWNT samples consist principally in achiral armchair tubes, of which, in these samples, the singular (10,10) tube is dominant.

Acknowledgements

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